



Eur pâisches
Patentamt

Eur pean
Patent Office

Office eur péen
des brevets

EP00/6203

REC'D 10/01/94 47
29 SEP 2000

WIPO

PCT

4

H5
4-2-03

Bescheinigung

Certificate

Attestation

Die angehefteten Unterlagen stimmen mit der ursprünglich eingereichten Fassung der auf dem nächsten Blatt bezeichneten europäischen Patentanmeldung überein.

The attached documents are exact copies of the European patent application described on the following page, as originally filed.

Les documents fixés à cette attestation sont conformes à la version initialement déposée de la demande de brevet européen spécifiée à la page suivante.

Patentanmeldung Nr. Patent application No. Demande de brevet n°

99112596.4

Der Präsident des Europäischen Patentamts;
Im Auftrag

For the President of the European Patent Office

Le Président de l'Office européen des brevets
p.o.

I.L.C. HATTEN-HECKMAN

**PRIORITY
DOCUMENT**
SUBMITTED OR TRANSMITTED IN
COMPLIANCE WITH RULE 17.1(a) OR (b)

DEN HAAG, DEN
THE HAGUE, 25/09/00
LA HAYE, LE





**Blatt 2 der Bescheinigung
Sheet 2 of the certificate
Page 2 de l'attestation**

Anmeldung Nr.:
Application no.: **99112596.4**
Demande n°:

Anmeldetag:
Date of filing: **01/07/99**
Date de dépôt:

Anmelder:
Applicant(s):
Demandeur(s):
Fraunhofer-Gesellschaft zur Förderung der angewandten Forschung e.V.
80636 München
GERMANY

Bezeichnung der Erfindung:
Title of the invention:
Titre de l'invention:
Polycyanate copolymers prepared from polyfunctional cyanates and fluorinated monocyanates

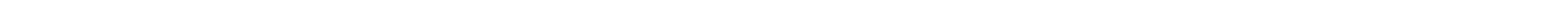
In Anspruch genommene Priorität(en) / Priority(ies) claimed / Priorité(s) revendiquée(s)

Staat: State: Pays:	Tag: Date: Date:	Aktenzeichen: File no. Numéro de dépôt:
---------------------------	------------------------	---

Internationale Patentklassifikation:
International Patent classification:
Classification internationale des brevets:
C08G73/06, G02B1/04

Am Anmeldetag benannte Vertragstaaten:
Contracting states designated at date of filing: AT/BE/CH/CY/DE/DK/ES/FI/FR/GB/GR/IE/IT/LI/LU/MC/NL/PT/SE
Etats contractants désignés lors du dépôt:

Bemerkungen:
Remarks:
Remarques:



01. Juli 1999

Polycyanate copolymers prepared from polyfunctional cyanates and fluorinated monocyanates

The present invention is directed to optical elements made of plastics and specifically those prepared by copolymerization of polyfunctional cyanates and fluorinated monocyanates, as well as to the said copolymers themselves.

Organic polymers are increasingly interesting materials in the optical or microoptical field, in integrated optics or in microsystem techniques. In these fields, they may be used in optical instruments and apparatuses or parts thereof as well as in special optics as lenses, prisms, for fixation of optical systems, as support material for optical layers and as translucent coating materials for mirrors or lenses. Organic polymers may be used in optical fibres and for the preparation of waveguide structures. Their technical handling is relatively easy, and their density is lower in comparison to glass.

Specifically, if such plastics or organic polymers are to be used as a waveguide, a variety of requirements are to be met. The refractive index of the material should be variable in a range as broad as possible and should be adaptable to that of specific substrates. If used in the optical communication engineering, low absorptions of the materials are required at 1.3 and 1.55 μ m. The loss due to attenuation caused by volume defects (non-homogenities, microbubbles, microfissures) should be minimized. Besides specific technological requirements, e.g. preparation of layers and structurability, specific provisions for the use of organic polymers as waveguide structures in integrated optics are the thermal and thermo-mechanical stability, adapted extension coefficients and long term stability of optical properties.

Until now, polymethacrylates and polycarbonates have been mainly used for optical purposes. Both classes of polymers have an excellent light transmittance, but their thermal and thermo-mechanical stability is not sufficient due to their chemical

structure. Thus, polymethacrylates and polycarbonates cannot practically be used at temperatures exceeding 130°C which is due to their relatively low glass transition temperatures. In addition, both types of polymers are linear, un-crosslinked polymers. This has the adverse effect that they are partly solubilized in case multilayer-systems are prepared via the application of dissolved components, e.g. by spin-coating each layer. Consequently, the layer structures as obtained are not sufficiently delimitated and neat which, however, is an essential for the preparation of waveguide structures.

There are other high performance polymers which have glass transition temperatures of more than 180°C. Examples are polyarylethersulfones, polyarylsulfones, polyaryletherketones, polyimides and polyetherimides, the processing of which, however, is more difficult than that of polymethacrylates and polycarbonates. Another disadvantage of these systems is the relatively high optical loss at wave lengths of 1.3 and 1.55µm, relevant in communication engineering.

20 Polyperfluorocyclobutanes (PFCB) are a relatively new class of high performance polymers. Upon thermal curing they yield unsoluble cross-linked polymers which are characterized by high thermal stability. Waveguide layers prepared from PFCB show very 25 low optical losses of 0.2 dB/cm at 1550 nm.

Also, polycyanurates have been used for the preparation of optical components. US patents 5 208 892 and 5 165 959 describe the preparation of polycyanate resins made of a single monomer (either fluorinated or non-fluorinated). German 30 Offenlegungsschrift DE 44 35 992 A1 describes optical elements prepared from polycyanurate resins. The resins are made by polymerization of dicyanate or polycyanate compounds, optionally in mixture with di- or polyphenols or di- or polyglycidyl 35 compounds. Like polyperfluorocyclobutanes, polycyanurates yield unsoluble cross-linked polymers upon thermal curing, and these polymers are as well characterized by high thermal stability. They are specifically useful due to their excellent adhesional

force on a variety of substrates, for example silicon, silica or a variety of organic polymers. Refractive index and glass transition temperature of the cured cross-linked polymers may be varied in broad ranges, due to the easy availability of a great number of di- and mono-functional cyanate monomers which may be copolymerized with each other. Polycyanurates of the kind mentioned above are partly commercially available. Completely cured polycyanurates known in the art which consequently are stable for long terms may have optical losses of about 0.2 dB/cm at 1.3 μ m. However, the optical losses are not less than 0.5dB/cm at 1.55 μ m which is important in communication engineering technologies.

It is the problem of the present invention to provide polymers which are thermally and thermomechanically stable for long terms, which may be easily compounded and manufactured and which have low optical losses at 1.3 and at 1.55 μ m, which are useful for the preparation of optical elements. Further, it is an object of the invention to provide optical elements and waveguide systems prepared from such polymers.

This problem has been solved by providing copolymers, obtainable by copolymerization of at least one monocyanate, derived from a partly or fully fluorinated alcohol ("fluorinated monocyanate"), and at least one polyfunctional organic cyanate ("polycyanate"). It has been found that such copolymers are specifically valuable in the preparation of optical elements, preferably of waveguides having the above mentioned desired properties.

Throughout the invention, "partly fluorinated" means that at least one fluorine atom is present in the molecule. "Fully fluorinated" means that hydrogen atoms are completely substituted by fluorine atoms. The whole molecules, or single organic radicals or groups (e.g. methyl, methylene, alkyl, aryl groups), respectively, may be fully fluorinated.

As fluorinated monocyanate, one, two, three or even more monocyanates of formula I may be used

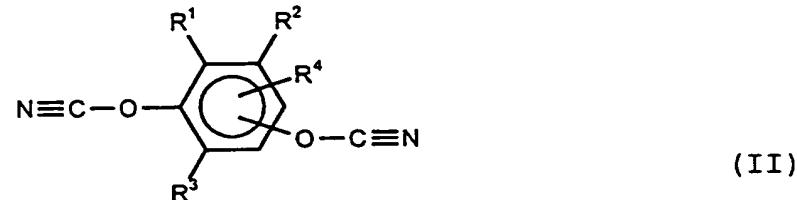
N≡C-O-R

(I)

wherein R is $C(R')_2-CFR''_2$, wherein each R' is, independently from
5 the other, hydrogen or fluorine or an optionally substituted,
preferably fluorinated alkyl or alkenyl group having preferably
1 to 13, more preferably 3 to 11 carbon atoms. Each of R'' may
independently be defined as R'. Further, R'' may have an aryllic
10 structure. Preferably, R is a straight, branched, or cyclic non-
aromatic hydrocarbon radical or an non-aromatic hydrocarbon
radical comprising a cyclic structure. Preferably, the non-
aromatic hydrocarbon radical has 1 to 15, more preferably 3 to
12 carbon atoms. It is to note that each of the carbon atoms of
15 R may carry 1, 2 or, if it is a terminal carbon atom, 3 fluorine
atoms. Fully fluorinated carbon atoms (-CF₃, -CF₂-) are
preferred. Further, it is preferred that one or both of R' are
hydrogen and/or one of R'' is fluorine or a partly or fully
fluorinated alkyl and the other is a partly or, more preferable,
fully fluorinated alkyl which may be straight, branched or
20 cyclic. Specific examples for the cyanates of formula (I) are
-CH₂-CF₂-CF₃, -CH₂-CF₂-CF₂-CF₃, -CH₂-C(CF₃)₂F, -CH₂-CF₂-CF₂-CF₃.

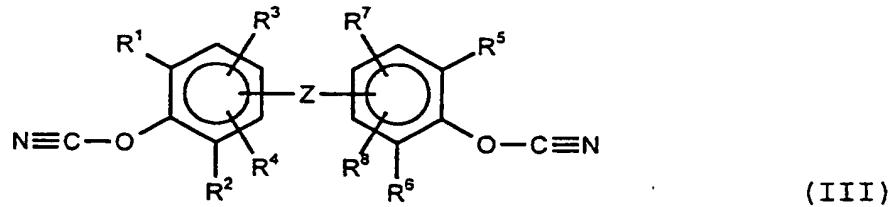
For the preparation of the said copolymer, one, two, three or
even more polyfunctional organic cyanates may be used. The
25 expression "polyfunctional" means that at least two NCO groups
are present in the molecule. However, the polyfunctional cyanate
may carry even more NCO groups, for example three, four or even
up to at least 22. The NCO groups are bound to organic radicals
30 via the oxygen atom. The polyfunctional cyanate may be, but is
not necessarily, partly or fully fluorinated. The organic
structure of the polyfunctional cyanate or cyanates is not
necessarily determined; however, the following polyfunctional
cyanates may preferably be selected under:

1. Difunctional cyanates of formula II:



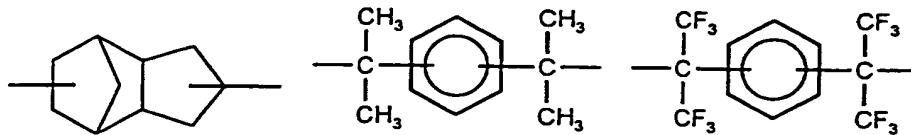
wherein R¹ to R⁴ are independently from each other hydrogen, optionally substituted C₁-C₁₀ alkyl, C₃-C₈ cycloalkyl, C₁-C₁₀ alkoxy, halogen, phenyl or phenoxy. The alkyl or aryl groups may be unfluorinated or they may carry at least one fluorine atom. In one embodiment, at least one of the alkyl or aryl groups present in the molecule is fully fluorinated. In another embodiment, the difunctional organic cyanate molecule is fully fluorinated. Examples are phenylene-1,3-dicyanate, phenylene-1,4-dicyanate, 2,4,5-trifluorophenylene-1,3-dicyanate or 2,3,5-trifluorophenylene-1,4-dicyanate.

2. Difunctional cyanates of formula III:



25

wherein R¹ to R⁴ and R⁵ to R⁸ are defined as R¹ to R⁴ above, Z is a chemical bond, SO₂, CF₂CH₂, CHF, CH(CH₃), isopropylene, hexafluoroisopropylene, n- or iso-C₁-C₁₀ alkylene which may be partly or fully fluorinated, O, NR⁹, N=N, CH=CH, C(O)O, CH=N, CH=N-N=CH, alkyloxyalkylene having 1 to 8 carbon atoms which is optionally partly or fully fluorinated, S, Si(CH₃)₂; R⁹ is hydrogen or C₁-C₁₀ alkyl, or

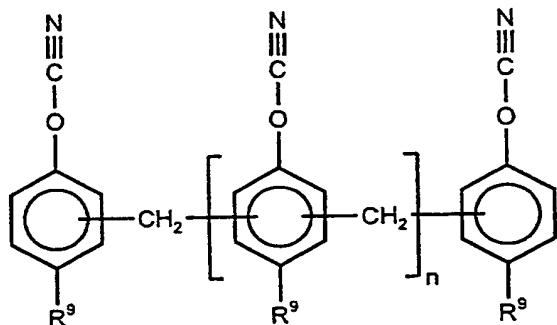


Examples are 2,2'-bis(4-cyanato-phenyl)propane,
2,2'-bis(4-cyanato-phenyl)hexafluoropropane,
biphenylene-4,4'dicyanate,
2,3,5,6,2',3',5',6'octafluorobiphenylene-4,4'-dicyanate.

5

3. Polyfunctional cyanates of formula IV:

10



(IV)

15 wherein R⁹ is defined as above and n is an integer of from 0 to 20.

20

4. Dicyanates according to formula V:



(V)

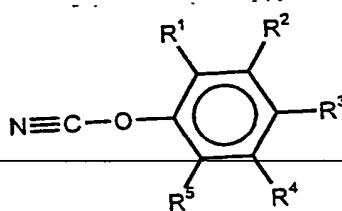
25

30

wherein R¹⁰ is an organic non-aromatic hydrocarbon group carrying at least 1 fluorine atom. Preferably, R¹⁰ is an alkylene group, more preferably having 3 to 12 carbon atoms. Each of the carbon atoms may carry 0, 1 or 2 or, in the case of a terminal group, 3 fluorine atoms. The carbon chain may be straight or branched or may be cyclic or may contain a cyclic part. Further, it may contain one or more C=C double bonds. In one embodiment, R¹⁰ is fully fluorinated. Examples are -CH₂-CF₂-CF₂-CH₂- or -CH₂-CF₂-CF₂-CF₂-CH₂.

In a preferred embodiment of the present invention, at least one additional monocyanate having formula VI:

35



(VI)

wherein R¹ to R⁵ are as previously defined for formulas II and III, is copolymerized in addition to the starting cyanates as defined above in order to obtain the polycyanate copolymer.

5 Examples for compounds of formula VI are phenylcyanate and perfluorophenylcyanate. Specifically, the refractive index and the glass transition temperature may be influenced by this additive as desired.

10 The polycyanate copolymers according to the invention may be obtained by mixing at least one of the monocyanates of formula I, optionally in addition to at least one of formula VI, and at least one polyfunctional organic cyanate, preferably at least one of the polycyanates of formula II to V. The ratio of 15 monocyanates to polycyanates may be freely chosen, provided that at least 1% by mol, preferably at least to 5% by mol, more preferably at least 10% by mol of monocyanate of structure I is present per mol of monomers to be polymerized. Preferably, the monofunctional cyanates of formulas I and VI are present in a 20 molar amount of not more than 75% related to the total amount of moles of monomers present in the mixture to be copolymerized.

The starting monocyanate and polycyanate compounds as described above are preferably warmed up after mixing. The temperature may 25 be chosen as required; a range of about 120°C to 170°C is preferred. Preferably, the reaction is performed in the absence of oxygen, e.g. in a sealed and preferably (under an inert gas atmosphere). The mixture is allowed to react until a liquid or viscous prepolymer (resin) is obtained. This prepolymer or resin 30 is soluble in useful solvents, preferably in solvents having high polarity, e.g. ethylethoxyacetate or chlorobenzene. In general, the prepolymer is processed in a respective solution, e.g. by spin-coating of a solution containing 25 to 65% by weight of the prepolymer, more preferably about 50% by weight of 35 the prepolymer. The prepolymer solution may be applied to a suitable substrate, consisting of e.g. silicon, quartz or an organic polymer. After being brought into the desired shape (e.g. a layer of desired thickness) it is cured (e.g. at

temperatures in the range of 200° to 260°C) in order to provide the desired network between the cyanate groups.

5 If an optical system comprising a variety of different layers of the present polycyanate copolymers shall be prepared, each different layer is applied and is cured, e.g. thermically cured, before the next layer is applied.

10 It shall be clear that the term "resin" is independent of the condition of the polymer, e.g. whether it is in a prepolymerized condition or is partly or completely cured.

15 The polycyanate copolymers according to the present invention have a glass transition temperature in the range of 100° to 300°C, and their refractive index at 1.55μm may be controlled in the desired range, specifically of from 1.35 to 1.60.

20 Specifically, the more fluorinated monomers are used, or the more fluorine parts per weight are present in the mixture, related to the weight of the mixture to be polymerized, the lower is the refractive index of the polycyanate copolymer obtained.

25 On the other hand, use of brominated derivatives of the cyanate monomers as defined above will raise the refractive index of the copolymer obtained. Thus, monocyanate compounds of e.g. formula VI wherein at least one of R¹, R², R³, R⁴ or R⁵ is substituted by bromine, may be advantageously added to the mixture. In general, the more bromine is included in the polymer, the higher is the refractive index obtained. Accordingly, any of the cyanates of 30 formulas I to VI as defined above (with the proviso that those of formula I may be free of fluorine) carrying one or more bromine atoms may be selected. However, brominated monocyanates are preferably used, either alone or in mixture with brominated polycyanates.

35 The polycyanate copolymers according to the present invention may be used for the preparation of optical elements. For example, they may be used for the preparation of waveguides and

waveguide structures. For such structures, use of at least two different polycyanate copolymers is preferred, wherein a polycyanate copolymer having a lower refractive index may be used for buffer and/or cladding while a polycyanate copolymer differing from the first one and having a greater refractive index may be used as the optical waveguide. At least one of these polycyanate copolymers should have been obtained according to the present invention. The selection will be easily made by a skilled person who is able to control the refractive index via the teachings given in this application. The layers show excellent adhesion to each other and to the substrate. Waveguide structures as described above may be prepared by known methods, e.g. RIE (Reactive Ion Etching).

The invention is now further illustrated by way of examples.

Example 1

5 12.9 g of a substituted dicyanate of Bisphenol A (compound III wherein R¹-R⁴ is H, R⁵-R⁸ is H, Z is hexafluoroisopropyl) and 3.7 g of a partly fluorinated monocyanate (compound I wherein R is CH₂-CF₂-CF₂-CF₃) are heated to 160°C in a sealed vessel for a time of about four hours. The reaction is terminated before gelling
10 starts, and a clear, pale yellow prepolymer is obtained which is viscous at 160°C and is solid at room temperature. The prepolymer is brought into solution by mixing it with 50 % by weight of ethylethoxyacetate (EEA). Spin-coating of this solution onto a substrate made of silicon wafer yields a layer
15 which may be cured at 240°C for one hour in a drying oven. The product has a refractive index of 1.4776 at 1.55 µm.

Example 2

20 12.9 g of a substituted dicyanate of Bisphenol A (compound III wherein R¹-R⁴ is H, R⁵-R⁸ is H, Z is hexafluoroisopropyl), 3.7 g of a partly fluorinated monocyanate (compound I wherein R is CH₂-CF₂-CF₂-CF₃), and 1.3 g of a monocyanate (compound VI wherein R¹, R², R⁴, R⁵ are hydrogen and R³ is bromine) are heated to 160°C in
25 a sealed vessel for a time of about four hours. The reaction is terminated before gelling starts, and a clear, pale yellow prepolymer is obtained which is viscous at 160°C and is solid at room temperature. The prepolymer is brought into solution by mixing it with 50 % by weight of EEA. Spin-coating of this solution onto a substrate made of silicon wafer yields a layer
30 which may be cured at 240°C for one hour in a drying oven. The product has a refractive index of 1.4870 at 1.55 µm.

Example 3

9.7 g of dicyanate of Bisphenol A (compound III wherein R¹-R⁴ is H, R⁵-R⁸ is H, Z is isopropyl) and 2.5 g of a fully fluorinated 5 monocyanate (compound I wherein R is C-(CF₃)₂) are heated to 140°C in a sealed vessel for a time of about four hours. The reaction is terminated before gelling starts, and a clear, pale yellow prepolymer is obtained which is viscous at 140°C and is solid at room temperature. The prepolymer is brought into 10 solution by mixing it with 50 % by weight of EEA. Spin-coating of this solution onto a substrate made of silicon wafer yields a layer which may be cured at 240°C for one hour in a drying oven. The product has a refractive index of 1.5596 at 1.55 µm.

15

Example 4

9.7 g of a substituted dicyanate of Bisphenol A (compound III wherein R¹-R⁴ is H, R⁵-R⁸ is H, Z is hexafluoroisopropyl), 10.3 g 20 of a partly fluorinated dicyanate (compound V wherein R¹⁰ is CH₂-CF₂-CF₂-CF₂-CH₂) and 1.1 g of a partly fluorinated monocyanate (compound I wherein R is CH₂-CF₂-CF₂-CF₃) are heated to 140°C in a sealed vessel for a time of about four hours. The reaction is 25 terminated before gelling starts, and a clear, pale yellow prepolymer is obtained which is viscous at 140°C and is solid at room temperature. The prepolymer is brought into solution by mixing it with 50 % by weight of EEA. Spin-coating of this solution onto a substrate made of silicon wafer yields a layer which is cured at 240°C for one hour in a drying oven. The product has a refractive index of 1.3689 at 1.55 µm.

Example 5

A 50 weight-% solution of the prepolymer of example 1 in EEA is spin-coated onto a silicon wafer, yielding a layer of about 8 μm thickness. Curing is performed at 240°C in a drying oven for one hour. Onto this layer, a 50 weight-% solution of the prepolymer of example 2 in EEA is spin-coated, again yielding a layer of about 8 μm thickness. Also, this layer is cured at 240°C in the drying oven for about 1 hour. According to known methods, an aluminum layer of about 100 nm is sputtered onto the said second prepolymer layer followed by its structurization by way of photolithography and chemical etching. Subsequently, the waveguides are structured by aid of oxygen RIE techniques (typical rate 100 nm/min using pure oxygen), and the etching mask is removed by treatment in a chemical etching bath. Then, the upper cladding layer is applied by spin-coating a prepolymer solution of example 1 followed by curing at 240°C for 1 hour. Using near field technique a difference of 0.0094 of the refractive index between the waveguide and its surrounding is

waveguides of different lenght yielded a loss of 0.35 dB/cm at 1.55 μm .

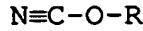
* * *

EPO-Munich
52

01. Juli 1999

Claims:

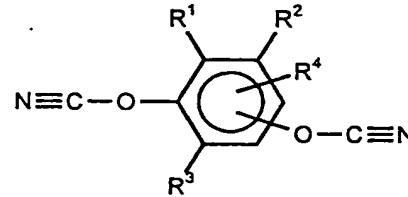
1. Polycyanate copolymer, obtainable by copolymerization of at least one polyfunctional cyanate with at least one monocyanate of the following formula I:



(I)

wherein R is a straight or branched non-aromatic hydrocarbon radical or a non-aromatic hydrocarbon radical comprising a cyclic structure, the radical having the formula $\text{C}(\text{R}')_2-\text{CFR}''_2$ wherein each R' is, independently from the other, hydrogen or fluorine or an optionally substituted, preferably fluorinated alkyl or alkenyl group, and each of R'' may independently be defined as R' or may have an arylic structure.

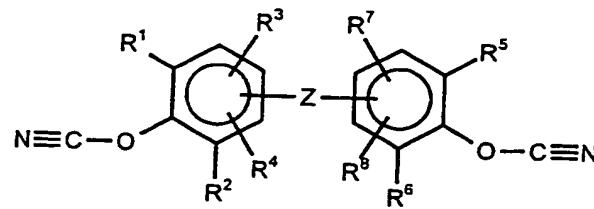
2. Polycyanate copolymer according to claim 1, wherein the polyfunctional cyanate(s) is/are selected from the group consisting of difunctional cyanates of formula II:



(II)

wherein R¹ to R⁴ are independently from each other hydrogen, optionally substituted $\text{C}_1\text{-C}_{10}$ alkyl, $\text{C}_3\text{-C}_8$ cycloalkyl, $\text{C}_1\text{-C}_{10}$ alkoxy, halogen, phenyl or phenoxy, the alkyl or aryl groups being unfluorinated, partly fluorinated or fully fluorinated,

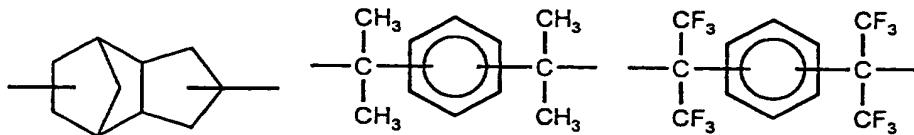
difunctional cyanates of formula III:



(III)

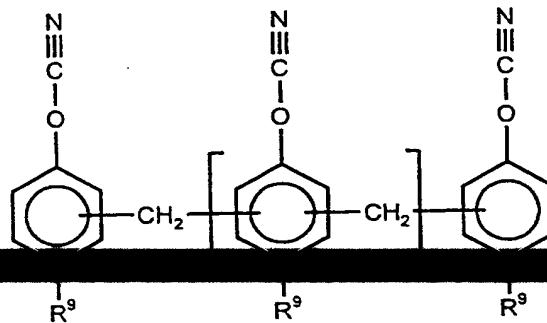
wherein R¹ to R⁴ and R⁵ to R⁸ are defined as R¹ to R⁴ above, Z is a chemical bond, SO₂, CF₂CH₂, CHF, CH(CH₃), isopropylene, hexafluoroisopropylene, n- or iso-C₁-C₁₀ alkylene, O, NR⁹, N=N, CH=CH, C(O)O, CH=N, CH=N-N=CH, alkyl oxyalkylene having 1 to 8 carbon atoms, S, Si(CH₃)₂, and R⁹ is hydrogen or C₁-C₁₀ alkyl or

10



polyfunctional cyanates of formula IV:

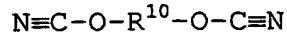
15



wherein R⁹ is as defined above and n is an integer of from 0 to 20, and

25

dicyanates having formula V:



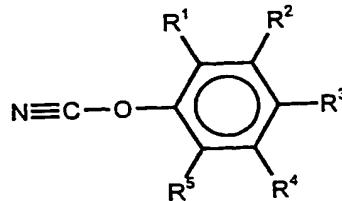
30

wherein R¹⁰ is a non-aromatic hydrocarbon group carrying at least one fluorine atom.

3. Polycyanate copolymer according to claim 2, wherein R¹⁰ of formula V is a partly or fully fluorinated alkylene group having 1 to 15, more preferably 3 to 12 carbon atoms.

4. Polycyanate copolymer according to any of the preceding claims, obtainable by copolymerization of a monocyanate of formula VI

5



wherein R¹ to R⁵ are defined as in the formulas II and III,
10 in addition to the cyanates as defined in any of claims 1 to 3.

15

5. Polycyanate copolymer according to any of the preceding claims, obtainable by copolymerization of at least one brominated monocyanate of formulas I to VI, preferably of formulas I or VI, as defined above with the proviso that the monocyanates of formula I may be free of fluorine, in addition to the cyanates as defined in any preceding claims.

20

6. Polycyanate copolymer according to any of the preceding claims, wherein the monocyanate of formula I as defined in claim 1 is used in an amount of at least 10%, preferably of at least 20% by mol per mol of the polycyanate copolymer.

25

7. Polycyanate copolymer according to claim 6, wherein the monocyanates of formulas I and VI as defined in claims 1 and 4, respectively, are used in an amount of up to 75% by mol per mol of the total monomers used for copolymerization.

8. Polycyanate copolymer according to any of the preceding claims, having a glass transition temperature of from 100°C to 300°C and/or having a refractive index of about 1.35 to about 1.60 at 1.55μm.

30 9. Polycyanate copolymer according to any of the preceding claims in a prepolymer condition.

10. Polycyanate copolymer according to any of claims 1 to 8 in a cured condition.

11. Optical element, consisting of or containing a resin composed of at least one polycyanate copolymer of any of 5 claims 1 to 8.

12. Optical element according to claim 11 which is an optical fibre or a waveguide.

13. Use of an optical element according to claim 11 or 12 in the optical or microoptical field, in integrated optics, or in 10 microsystems.

14. Use of an optical element according to claim 11 as a lens, a prism, for fixation of optical systems, as a support for optical layers, or as a translucent coating material for mirrors or lenses.

consisting of a resin composed of one or more of a polycyanate copolymer of claims 1 to 8, and a buffer and/or cladding, consisting of a resin composed of one or more of a polycyanate copolymer of any of claims 1 to 8 different from 20 that of the waveguide, wherein the resin of the waveguide has a greater refractive index than that of the buffer and/or cladding.

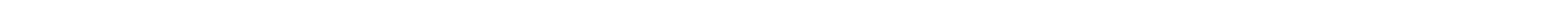
16. Method for producing a polycyanate copolymer according to any of claims 1 to 8 in a prepolymer condition, comprising 25 the following steps:

mixing at least one polycyanate, preferably as defined in claim 2 or 3, and at least one monocyanate of formula I as defined in claim 1, and warming the mixture in a sealed vessel to a temperature at which a prepolymer forms.

17. Method for producing a polycyanate copolymer according to any of claims 1 to 8 in a cured condition, comprising the following steps:

mixing at least one polycyanate, preferably as defined in claim 2 or 3, and at least one monocyanate of formula I as defined in claim 1, and warming the mixture in a sealed vessel to a temperature at which a prepolymer forms, dissolving the prepolymer in a suitable solvent, applying the dissolved prepolymer in the desired shape and curing the prepolymer at a temperature at which crosslinking occurs.

* * *



01 Juli 1999

Polycyanate copolymers prepared from polyfunctional cyanates and fluorinated monocyanates

The present invention is directed to polycyanate copolymers, obtainable by copolymerization of at least one polyfunctional cyanate with at least one monocyanate of the formula $N\equiv C-O-R$, wherein R is a straight or branched non-aromatic hydrocarbon radical or a non-aromatic hydrocarbon radical comprising a cyclic structure, the radical having the formula $C(R')_2-CFR''_2$, wherein each R' is, independently from the other, hydrogen or fluorine or an optically substituted, preferably fluorinated alkyl or alkenyl group, and each of R'' may independently be defined as R' or may have an aryllic structure. Preferably, the at least one polyfunctional cyanate is selected from aliphatic or aromatic dicyanates or from oligocyanates comprising a straight chain of 2 to 22 oligomerized monocyanates. The polycyanate copolymer may further comprise an aromatic monocyanate and/or one or more of brominated cyanates. The polycyanate copolymers are advantageously used for the preparation of optical systems, e.g. waveguide structures, because they have low optical losses at 1.3 and 1.55 μ m.

* * *

